

Double-exchange is not the cause of ferromagnetism in doped manganites

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The coexistence of ferromagnetism and metallic conduction in doped manganites has long been explained by a double-exchange model in which the ferromagnetic exchange arises from the carrier hopping. We evaluate the zero-temperature spin stiffness $D(0)$ and the Curie temperature T_C on the basis of the double-exchange model using the measured values of the bare bandwidth W and the Hund's rule coupling J_H . The calculated $D(0)$ and T_C values are too small compared with the observed ones even in the absence of interactions. A realistic onsite interorbital Coulomb repulsion can reduce $D(0)$ substantially in the case of a 2-orbital model. Furthermore, experiment shows that $D(0)$ is simply proportional to x in $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ system, independent of whether the ground state is a ferromagnetic insulator or metal. These results strongly suggest that the ferromagnetism in manganites does not originate from the double-exchange interaction. On the other hand, an alternative model based on the $d-p$ exchange can semi-quantitatively explain the ferromagnetism of doped manganites at low temperatures.

The discovery of “colossal” magnetoresistance in thin films of the manganite perovskites $\text{Re}_{1-x}\text{D}_x\text{MnO}_3$ (Re = a rare-earth element, and D = a divalent element) [1,2] has stimulated extensive studies of magnetic, structural and transport properties of these materials [3]. The coexistence of ferromagnetism and metallic conduction has long been explained by the double-exchange (DE) model [4,5], where the effective hopping for the manganese $3d$ conduction electrons varies with the angle between the manganese core electrons due to a strong Hund's coupling. However, Millis *et al.* [6] proposed that, in addition to the double-exchange, a strong electron-phonon interaction arising from a strong Jahn-Teller effect should be involved to explain the basic physics of manganites. In this modified model, the primary cause of the ferromagnetism of doped manganites is still the double-exchange interaction.

In the DE model, it is implicitly assumed that doped carriers are $\text{Mn } e_g$ electrons. This assumption is not justified by both electron-energy-loss [7] and photoemission spectroscopies [8], which have shown that the ferromagnetic manganites ($x < 0.4$) are doped charge-transfer insulators with carriers mainly residing on the oxygen orbitals. Now a question arises: Does the ferromagnetism of doped manganites really originate from the DE interaction? If not, what causes the ferromagnetism in these compounds? One way to address this fundamental issue is to make a quantitative comparison between the predicted properties of the DE model and experiment.

There are two important parameters in the DE model, namely, the bare bandwidth W of the e_g bands, and the Hund's rule coupling J_H between e_g and t_{2g} electrons. These parameters are related to an optical transition between the exchange splitted e_g bands [9,10], and thus can be determined from optical data. With these unbi-

ased parameters, one can calculate the zero-temperature spin stiffness $D(0)$ and T_C . Here $D(0)$ is defined as $\omega_q = D(0)q^2$ with ω_q being the magnon frequency. When one introduces interactions such as electron-phonon and electron-electron interactions, the magnitudes of both $D(0)$ and T_C are generally reduced.

Here we use the measured values of the bare bandwidth W and the Hund's rule coupling J_H to calculate $D(0)$ and T_C on the basis of the double-exchange model. The calculated $D(0)$ and T_C values are too small compared with the observed ones. A realistic onsite interorbital Coulomb repulsion can reduce $D(0)$ substantially in the case of a 2-orbital model. Moreover, experimental data show that $D(0)$ is simply proportional to x in $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ system, independent of whether the ground state is a ferromagnetic insulator or metal. These results provide strong evidence that the ferromagnetism in manganites is not caused by the DE interaction. On the other hand, an alternative model based on the $d-p$ exchange can well explain the ferromagnetism of doped manganites.

Now we start with a Kondo-lattice type Hamiltonian [11], which leads to Zener's DE model when $J_H \rightarrow \infty$,

$$H = -\frac{1}{2} \sum_{\langle ij \rangle a b \alpha} t_{ij}^{ab} (d_{ia\alpha}^\dagger d_{jb\alpha} + h.c.) - J_H \sum_{i\alpha\alpha\beta} \vec{S}_i \cdot d_{ia\alpha}^\dagger \vec{\sigma}_{\alpha\beta} d_{i\alpha\beta} + H_{INT}. \quad (1)$$

Here $d_{ia\alpha}^\dagger$ creates an electron in e_g orbital a with spin α , t_{ij}^{ab} is the direction-dependent amplitude for an electron to hop from orbital a to orbital b on a neighboring site, and H_{INT} represents the other interactions. The calculated band structure is well fit by a t_{ij}^{ab} , which involves only nearest-neighbor hopping that is only nonzero for

one particular linear combination of orbitals, i.e., $t_{ij}^{ab} \propto t$, where t is a characteristic hopping amplitude that is related to the bare bandwidth W by $W = 4t$. Here we still call Eq. 1 as 2-orbital DE model rather than 2-orbital Kondo-lattice model for convenience. The quantum and thermal average of the hopping term in Eq. 1, defines a quantity K :

$$K = (1/6N_{site}) \sum_{\langle ij \rangle ab\alpha} t_{ij}^{ab} \langle d_{ia\alpha}^\dagger d_{jb\alpha} + h.c. \rangle, \quad (2)$$

The quantity K is related to the optical spectral weight by a familiar sum rule,

$$K = \frac{2a_o}{\pi e^2} \int_0^\infty d\omega \sigma_1(\omega), \quad (3)$$

where σ_1 is the real-part optical conductivity contributed only from the e_g electrons, and a_o is the lattice constant. The quantity K generally consists of the Drude part K_D , and incoherent part K_I which, in general, involves inter-band and intraband optical transitions. The Drude part K_D can be related to the plasma frequency Ω_p as

$$K_D = \frac{a_o}{4\pi e^2} (\hbar \Omega_p)^2. \quad (4)$$

On the basis of Eq. 1, Quijada *et al.* [11] showed that, to the order of $1/J_H$, the spin stiffness $D(0)$ is,

$$D(0) = \frac{K a_o^2}{4S^*} \left[1 - \frac{\eta t^2}{J_H S K} \right], \quad (5)$$

where $S = 3/2$, $S^* = S + (1-x)/2$, and $\eta = 1.04$ when $H_{INT} = 0$. The presence of interactions may change the value of η . A similar result was obtained by Furukawa [9] for an 1-orbital DE model using the dynamical mean field method, but the value of η is doping dependent and less than 1.

From the above equations, one can calculate K and $D(0)$ using realistic values of the bare bandwidth W and the Hund's rule coupling J_H . Both the local density approximation (LDA) [12] and "constrained" LDA [13] calculations show that $J_H \simeq 1.5$ eV. The calculated J_H value is very close to the atomic values for 3d atoms. This is reasonable because J_H is not screened when the ion is put in a solid. The bare bandwidth W cannot be calculated reliably for 3d-metal based compounds due to a strong correlation effect. Fortunately, the values of both W and J_H can be determined from an optical transition between the exchange splitted e_g bands [9,10]. The peak position of this optical transition is about $2J_H$, and the width of the peak contains information about the bare bandwidth [9,10]. From the optical data of Ref. [11,14], one finds $J_H \simeq 1.6$ eV and $W = 1.6$ - 1.8 eV by comparing the data with the theoretical predictions [9,10]. The value of J_H obtained from the optical data is in excellent agreement with the calculated one. This implies that the

feature appeared at about 3 eV in the optical data indeed arises from the optical transition between the exchange splitted e_g bands.

The quantity K° for noninteracting 2-orbital model can be evaluated when the bare bandwidth W is known. Takahashi and Shiba [15] have calculated the optical conductivity using a tight binding (TB) approximation of the band structure. From their calculated result for the interband optical conductivity, we evaluate that $K_I^\circ = 0.088t$ for $x \sim 0.3$. One should also note that the magnitude of t defined in Ref. [15] is 1.5 times smaller than the t defined here. Since $K_D^\circ = 1.2K_I^\circ$ [15], then $K_D^\circ = 0.106t$ and $K^\circ = 0.194t$. The LDA calculation for a cubic and undistorted structure shows that [16] $W = 3$ eV and $\hbar\Omega_p = 1.9$ eV. Using Eq. 4 and $\hbar\Omega_p = 1.9$ eV, one yields $K_D^\circ = 78.6$ meV. Since $t = W/4 = 0.75$ eV, one readily finds that $K_D^\circ = 0.105t$, in remarkably good agreement with that ($K_D^\circ = 0.106t$) estimated from the TB approximation. This justifies the relation $K^\circ = 0.194t$ obtained from the TB approximation. It is interesting to compare the present result with those reported in Ref. [10] and [11]. In Ref. [10], it is found that $K^\circ = 0.34t$ for $x = 0.3$ using the dynamic mean field method. In Ref. [11], Quijada *et al.*, claimed $K^\circ = 0.46t$ for $x = 0.3$, which might be true if the two e_g bands have the same dispersion. Therefore the quantity K° is significantly overestimated in Ref. [10] and [11].

When the Hund's coupling J_H is turned on, the quantity K is reduced compared with K° . For $J_H = \infty$ and $x = 0.3$, $K = 0.77K^\circ$ [10,17]. It was also shown that [10] the reduction factor (0.77) is basically the same for $J_H \geq t$. Therefore, we have $K = 0.147t$ for $x = 0.3$. Using $t = 0.4$ eV, $J_H = 1.6$ eV, and $K = 0.147t$, we yield $D(0) = -25$ mV \AA^2 from Eq. 5. The negative value of $D(0)$ implies that the ferromagnetism is not sustainable with these unbiased parameters. It might be possible to have a small positive $D(0)$ if one includes higher order terms in Eq. 5. Nevertheless, the theoretical $D(0)$ value is too small compared with the measured ones (160-190 mV \AA^2) [18-20].

Now we turn to the calculation of T_C for $x = 0.3$. For an 1-orbital DE model with $J_H = \infty$, the dynamic mean field (DMF) calculation shows that $T_C^{MF} = 0.078t/k_B$ [9], while Monte Carlo simulations yield $T_C = 0.04t/k_B$ [21], or $T_C = 0.03t/k_B$ [22]. This implies that the DMF calculation overestimates T_C by a factor of about 2 due to the neglect of fluctuations. We would like to mention that the magnitude of t defined in Ref. [21] and [22] is 3 times smaller than the t defined here. For realistic parameters $t = 0.4$ eV, $J_H = 4t = 1.6$ eV, the DMF calculation obtains $T_C^{MF} = 0.038t/k_B = 180$ K [9]. Considering the fact that the DMF method can overestimate T_C by a factor of 2, one has $T_C \sim 100$ K. For the 2-orbital DE model, the DMF calculation shows [10] $T_C^{MF} = 0.07t/k_B = 324$ K. Since the quantity K calculated in Ref. [10] is overesti-

ated by a factor of about 2 (as discussed above), and the DMF method itself can overestimate T_C by about 2 times, the real T_C should be about 100 K, which is comparable with the value for the 1-orbital model. This is reasonable because the bare values of K° for both 1- and 2-orbital models happen to be very similar in the case of $x = 0.3$. Therefore, both the 1- and 2-orbital DE models cannot explain the observed T_C values with the unbiased parameters.

The above calculations have not taken into account any other interactions such as electron-phonon interaction and electron-electron correlation. The electron-phonon interaction can substantially reduce the K and thus $D(0)$ if the coupling constant $\lambda = E_p/2t$ is greatly larger than 1 [23,24]. In reality, the polaron binding energy E_p in manganites is estimated to be about 1 eV [25]. So $\lambda \simeq 1$, which suggests that the electron-phonon coupling does not lead to a sizable decrease in $D(0)$. On the other hand, the electron-electron correlation in the 2-orbital model can lead to a large reduction in K , as demonstrated by Horsch and coworkers [26]. The parameter $U = U' - J_{ab}$ (where U' is the onsite interorbital Coulomb repulsion, and J_{ab} is the interorbital Hund's coupling) has a strong influence on the value of K [26]. With realistic values of $U = 3$ eV [26,27] and $t = 0.4$ eV, K can be reduced by a factor of about 2 compared with the bare one [26]. Although their results are for 2-dimensional finite clusters, we would expect a similar result for a real 3-dimensional system.

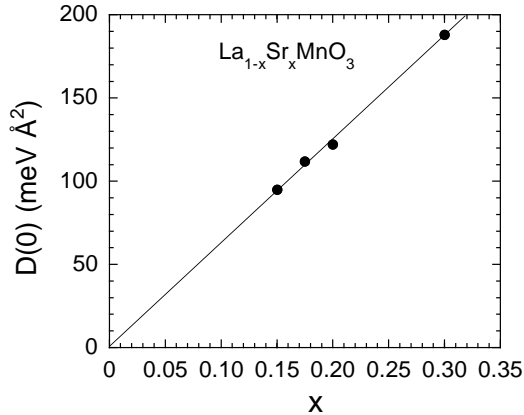


FIG. 1. Zero-temperature spin stiffness $D(0)$ vs x in $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ system. The $D(0)$ value for $x = 0.15$ is taken from Ref. [28], and the other data from Ref. [29]. It is evident that $D(0)$ is simply proportional to x , independent of whether the ground state is a ferromagnetic insulator (e.g., $x = 0.15$), or a ferromagnetic metal (e.g., $x = 0.175, 0.20, 0.30$).

In addition, it is striking that the observed spin stiffness varies only with x , namely, $D(0) \propto x$, as seen clearly from Fig. 1. The doping dependence of $D(0)$ shown in Fig. 1 could be qualitatively explained by the 1-orbital DE model [9,30] where $D(0) \propto K \propto x(1-x)$ for $J_H = \infty$

[30], as plotted in Fig. 2. Nevertheless, the doping dependence of $D(0)$ is very different from that predicted from the 2-orbital model. By analogy to the 1-orbital model, one can easily show that $D(0) \propto K \propto (1-x)(1+x)$ for the 2-orbital model, as demonstrated in Fig. 2. Here we have implicitly assumed that the two e_g bands have the same dispersion. The solid circles in Fig. 2 are the results evaluated using the dynamical mean field method with $J_H = \infty$ [10]. It is evident that the analytical expression is in excellent agreement with the numerical result. Comparing the results shown in Fig. 1 and Fig. 2, one clearly sees that the 2-orbital model cannot explain the observed doping dependence of $D(0)$.

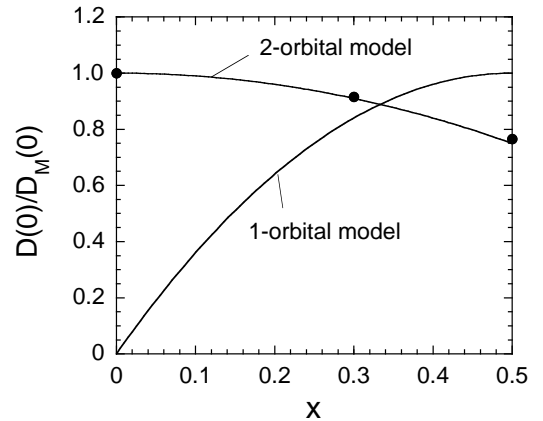


FIG. 2. The doping dependence of $D(0)$ predicted by the 1-orbital and 2-orbital DE models with $J_H \rightarrow \infty$. The solid lines represent $D(0) \propto x(1-x)$ for the 1-orbital model, and $D(0) \propto (1-x)(1+x)$ for the 2-orbital model. The solid circles are the values numerically calculated from the 2-orbital model [10].

The question is why the DE model cannot explain the ferromagnetism in doped manganites. As mentioned above, the DE model implicitly assumes that doped carriers are Mn e_g electrons, which is not the case according to the electron-energy-loss and photoemission spectra [7,8]. Furthermore, the “constrained” LDA calculation [13] shows a large onsite Coulomb repulsion of about 8-10 eV, in agreement with the photoemission data [8]. The simple LDA calculation which ignores the strong correlation effect shows a bare plasma frequency of about 1.3 eV for $x = 0.33$ with a distorted structure determined by neutron scattering [31]. The bare plasma frequency calculated is much smaller than the one observed in $\text{Nd}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ (3.3 eV) [32]. The large bare plasma frequency observed in this material is consistent with the fact that doped holes reside mainly on the oxygen orbitals with a large bandwidth. The bare plasma frequency of

about 3.3 eV for single conduction band (oxygen band) implies a bare K° of about 0.24 eV, which gives an upper limit for the K in the presence of interactions. The electron-phonon interaction with a coupling constant $\lambda \sim 1$ will reduce the K slightly [23,24], but can significantly decrease the Drude weight. Optical data indeed show that the K for $\text{Nd}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ is about 0.2 eV [11], while the effective plasma frequency is about 0.57 eV [33]. The Drude weight is reduced by a factor of 33, implying small polaronic carriers in the low-temperature ferromagnetic state.

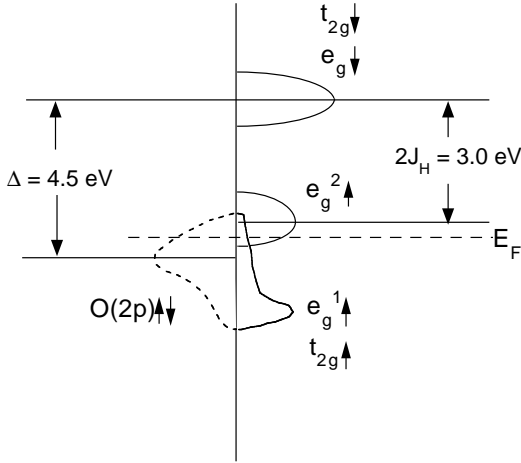


FIG. 3. The schematic band structure for doped manganites ($x < 0.4$) constructed from the LDA + U calculation [13]. The energy scales are consistent with the optical data [11,33,37]. The density of the oxygen holes is equal to x per cell while the densities of the Mn^{2+} and Mn^{4+} ions are the same due to the charge disproportion ($2\text{Mn}^{3+} \rightarrow \text{Mn}^{2+} + \text{Mn}^{4+}$) [36].

In Fig. 3, we plot a schematic band structure for doped manganites ($x < 0.4$), which is extracted from the LDA + U calculation. Here we have assumed that the local Jahn-Teller distortions still survive upon doping, but the average magnitude of the distortions decreases, in agreement with the experiments [34,35]. The doping with a divalent element shifts down the e_g^2 band due to the decrease of the Jahn-Teller distortions. The density of the oxygen holes is equal to x per cell, while the electron carrier density in the majority e_g^2 band (corresponding to the density of the Mn^{2+} ions) is the same as the hole carrier density in the majority e_g^1 band (corresponding to the density of the Mn^{4+} ions). In other words, the doping does not change the average valence of the Mn ions, but lead to the charge disproportion ($2\text{Mn}^{3+} \rightarrow \text{Mn}^{2+} + \text{Mn}^{4+}$). This is because the quench of the static Jahn-Teller distortions by doping makes the Mn^{3+} - Mn^{4+} pairs more stable than the Mn^{3+} - Mn^{3+} pairs [36]. The current band structure is consistent with the optical transitions at the photon energies of about 1.5 eV, 3.0 eV and 4.5 eV [11,33,37]. The optical spectral weight for the 4.5

eV transition should be much larger than for the 1.5 eV transition, as observed [37]. This is because the unoccupied state density for the former optical transition (i.e., 3 minority t_{2g} and 2 minority e_g states per cell) is at least 5 times larger than that for the latter one (i.e., less than 1 majority e_g states per cell). The optical transition at about 3 eV is related to the transition between the exchange splitted e_g bands.

What is an alternative model for the ferromagnetism in doped manganites? If we consider an oxygen hole (spin 1/2) sitting in between two Mn ions, an exchange interaction between the oxygen and Mn spins ($d-p$ exchange) will lead to a ferromagnetic interaction between Mn spins [38]. In this case, the ferromagnetic exchange energy between two Mn spins is $J = C(t_{pd}^4/\Delta_{ct}^3)$, where C is a numerical factor of order of 1, t_{pd} is a hybridization matrix element between the d and p orbitals, and Δ_{ct} is a charge transfer gap. The long-wave spin stiffness for a cubic material is [39] $D(0) = (S^*/3) \sum_j J_{ij} |\vec{R}_i - \vec{R}_j|^2$, where J_{ij} is the exchange energy between pairs of spins at sites \vec{R}_i and \vec{R}_j , and the sum is over six neighboring sites. Clearly, $J_{ij} = J$ if there is an oxygen hole in between two Mn ions, while $J_{ij} = 0$ if not. Since the oxygen hole density is equal to x per cell, then $D(0) = (2/3)xJS^*a_o^2$, independent of whether the holes are localized or not. This mechanism can naturally explain why $D(0)$ is simply proportional to x , as shown in Fig. 1. Using the Slater-Koster parameter $V_{pd\sigma} = 1.8$ eV obtained from the analysis of photoemission data [8], or from the TB fit to the LDA band structure [40], we yield $t_{pd} = -(\sqrt{3}/2)V_{pd\sigma} = -1.56$ eV [41]. Taking $\Delta_{ct} = 4.5$ eV [8,11,37], $t_{pd} = -1.56$ eV, $C = 1$, and $x = 0.3$, we obtain $D(0) = 0.5$ eV \AA^2 , which is larger than the observed values by about three times. This is not unreasonable since the numerical factor C might be less than 1. A more quantitative calculation of J based on the $d-p$ exchange is essential to address this issue.

It should be noted that the ferromagnetic “bonds” with the exchange energy J will be randomly distributed over the real space if the oxygen holes are localized, whereas the distribution of the bonds will become more homogeneous when the holes are more mobile (a motion narrowing effect). This can account for a conventional spin-wave dispersion in high T_C materials [39] where the conductivity is high, and an unconventional magnon softening near the zone boundary for lower T_C compounds [19] where the conductivity is lower. When the oxygen holes are ordered (charge-ordering), as observed in $\text{La}_{0.85}\text{Sr}_{0.15}\text{MnO}_3$ [28], the ferromagnetic bonds will form a “superlattice”, leading to a splitting of spin-wave dispersion at a wave vector equal to the incommensurability of the superlattice as observed [28].

The above simple model would imply that the Curie temperature T_C should be proportional to x . In reality, T_C is strongly dependent on the cation radius r_A of the

$\text{Re}_{1-x}\text{D}_x$ site of the perovskite $\text{Re}_{1-x}\text{D}_x\text{MnO}_3$ even if x is fixed [42]. Moreover, a giant oxygen isotope shift of T_C has been observed in these compounds [43,44]. Therefore, the simple $d-p$ exchange model cannot account for these unusual phenomena. Recently, Alexandrov and Bratkovsky [45] have proposed that, in addition to the $d-p$ exchange interaction, there is a strong electron-phonon interaction that may lead to the formation of small (bi)polarons. In the paramagnetic state, the singlet bipolarons (spin zero) are stable and the ferromagnetic interaction is produced by the thermally excited polarons (spin 1/2). Thus, the Curie temperature T_C is selfconsistently determined by the polaron density at T_C . Within this scenario, T_C can strongly depend on the electron-phonon coupling strength and the isotope mass [45], in agreement with experiment [43,44]. At zero temperature, all the carriers will be polarons if $J_{pd}S$ is much larger than the bipolaron binding energy Δ , where J_{pd} is the exchange energy between Mn and oxygen-hole spins. In this case, $D(0) \propto x$, as in $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ system. On the other hand, there is a mixture of polarons and bipolarons if $J_{pd}S$ is slightly larger than Δ . Then the relation $D(0) \propto x$ does not hold, and $D(0)$ is proportional to the density of polarons which is less than x per cell. One should also expect that the mixture of mobile polarons and localized bipolarons would lead to a dynamic/static phase separation since bosonic and fermionic carriers tend to separate in real space.

Finally we would like to address whether the band structure shown in Fig. 3 can be consistent with an effective 1-orbital DE model with a large $t \sim 1.2$ eV. We can show that in order to have a T_C of 380 K with such a t , one needs an unphysically large value of J_H , i.e., $J_H > 5$ eV. Since the $d-p$ exchange model can well explain the ferromagnetism, it would not be necessary to have such an effective DE model for describing the physics of manganites. We believe that the $d-p$ exchange + (bi)polaron model [45] is sufficient to account for the essential physics in manganites.

Acknowledgement: We would like to thank A. S. Alexandrov, S. Satpathy, A. Chattopadhyay, A. J. Millis, G. Khaliullin, E. Dagotto, K. A. Müller, H. Keller, T. Moritomo, T. A. Kaplan and P. C. Dai for useful discussions. The work was supported by the Swiss National Science Foundation.

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